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Second harmonic generation efficiency in KDP crystals containing alkali halides

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Abstract : Pure KDP and KDP crystals containing alkali halides with and without Au^{+} ions were grown from aqueous solution by slow evaporation. Second harmonic generation (SHG) efficiency measurements were carried out by Kurtz method. The present study justifies that the KDP crystals containing alkali halides have appreciable increase in SHG efficiency compared to pure KDP crystals.

Key words : Nonlinear optics, KDP crystals, second harmonic generation efficiency

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Potassium dihydrogen phosphate (KH_2PO_4 or KDP) and its isomorphs are representative of hydrogen bonded materials which possess important piezoelectric, electro-optic and nonlinear optical (NLO) properties [1,2]. They have attracted the interests of many theoretical and experimental researchers, probably because of their comparatively simple structure and very fascinating properties associated with hydrogen bond system involving large isotope effect. A most conspicuous feature in the development of physics of these crystals is the close interplay between theory and experiments, which make significant progress in the understanding of their microscopic properties possible.

For short pulse applications, a second order nonlinear material ideally must have a large threshold and a large nonlinear coefficient [3]. Among a variety of nonlinear crystals, potassium dihydrogen phosphate (KDP), Lithium triborate (LBO) and ß-barium borate (BBO) are the most widely used materials for second harmonic generation [4]. These inorganic nonlinear crystals have large damage thresholds and perform exceptionally well in most ultrashort-pulse applications [5]. These typically involve the generation of harmonics of Nd-based near infrared solid state lasers [6]. For example the 1064 nm fundamental of a Nd-YAG laser can be converted to its 532 nm second harmonic or to its 355 nm third harmonic or to its 266 nm fourth harmonic by using KDP.

Laser radiation in the near IR with high average power and good beam quality has become available mainly with the advent of advanced solid-state laser technology. Crystals with high conversion efficiencies for second harmonic generation are desirable in various fields. With the aim of discovering new useful materials for academic and industrial use, we have made an attempt to measure NLO activity in KDP crystals by means of adding alkali halides in definite ratios and doping with gold. In this paper, we are reporting for the first time the second harmonic generation efficiency in doped KDP crystals with Au^{+} ions as well as with alkali halides as additives.

The samples used in the present study were grown from aqueous solution at room temperature in the unstirred condition. KDP was added with the alkali halides (KCl/NaCl/KBr/NaBr/KI/NaI) in molecular ratio, KDP : alkali halides ; 9 : 1. Analytical reagent (AR) grade samples of KDP and alkali halides were used. Both the salts were taken (in

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their molar mass) separately and then mixed together. Supersaturated solutions of the mixed salts were kept for slow evaporation in beakers covered with filter paper at room temperature.

Au^+- doped crystals were grown by adding definite volume of tetra-chloro auric acid solution to the mixture of KDP and alkali halides to know the effect of metal ions on laser efficiency. All the grown crystals were found to be very stable, colourless and transparent. Scalenohedral (twelve sided polyhedron) morphology was exhibited by all the crystals grown.

The elemental analyses of the grown crystals were carried out by an energy dispersive X-ray spectroscopy (EDAX). It is observed from the EDAX data that the impurities have entered into the lattice of KDP crystals.

To evaluate conversion efficiency, Kurtz powder method [7] is an important tool for scientists searching for efficient NLO materials. The experimental setup used in the present investigation was similar to the generic one devised by Kurtz and is illustrated in Figure 1. It consisted of a Q-switched Nd : YAG laser, the output of which was filtered through 1064 nm narrow pass filter.

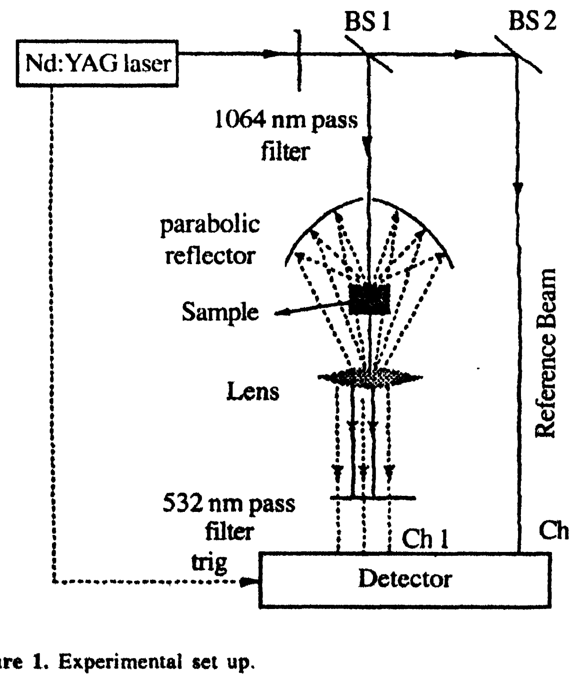

Figure 1. Experimental set up.

The power of the fundamental beam was monitored by a split beam technique, in one channel of the power meter. The sample was ground in the form of the fine powder of known grain size (30–50 Mm) and pressed between two glass plates. The sample size was kept larger than the beam cross section. The generated harmonic was passed through a 532 nm narrow pass filter and fed to the other channel of the power meter. The ratio of the fundamental and harmonic intensities gives the efficiency of the sample. To eliminate the experimental error, a urea sample of the same grain size was also tested in the same setup and the efficiency was evaluated as a ratio.

To measure the powder SHG efficiency, samples were derived from the crystals grown with full morphology which ensures homogeneity of the material. A Q-switched Nd-YAG laser whose output was filtered through 1064 nm narrow pass filter was used for this purpose. The input power of the laser beam was measured to be 16.5 mJ/pulse. Pure KDP was used as reference sample. Both the reference and test samples had uniform particle size (30–50 Mm)

The experiment was first carried out in pure KDP. Later in all the other samples namely KDP with alkali halides with and without Au^+ ions. Throughout the experiment the laser power was kept constant.

The results showed that the SHG efficiency of KDP containing alkali halides is higher than pure KDP. For NLO systems to show second order nonlinear activity, the additives and the dopants in them have to be macroscopically aligned, and then only there can be increase in efficiency, which has been justified experimentally [8]. The measured SHG signal and efficiency are given in the following table 1. The SHG signal $I_{2\omega}$ observed under the same experimental conditions for powder samples of KDP, KDP + alkali halides (KCl/KBr/NaBr/KI/NaI) with and without Au^+ ions revealed that $I_{2\omega}$ KDP $< I_{2\omega}$ KDP + alkali halides + $Au^+ < I_{2\omega}$ KDP + alkali halides. Only in case of NaCl, it has revealed $I_{2\omega}$ KDP $< I_{2\omega}$ KDP + NaCl $< I_{2\omega}$ KDP + NaCl + Au^+. However, final efficiency is due to the presence of Au^+ ions and additives. Enhancement or lowering of efficiency due to Au^+ ions gives an idea regarding better alignment or lower alignment of dopants in the crystal matrix. It is difficult to draw definite conclusion based on this result and it requires further work on phase matched SHG efficiency of single crystal.

Table 1. SHG efficiency.

Sample	SHG signal 2ω (mv)	Efficiency with respect to KDP
Pure KDP	16	1.00
Au^+ doped KDP	17	1.0625
KDP + KCl	20	1.25
Au^+ doped KDP + KCl	18	1.125
KDP + NaCl	17	1.0625
Au^+ doped KDP + NaCl	22	1.375
KDP + KBr	17	1.0625
Au^+ doped KDP + KBr	18	1.125
KDP + NaBr	19	1.1875
Au^+ doped KDP + NaBr	17	1.0625
KDP + KI	17	1.0625
Au^+ doped KDP + KI	16	1.00
KDP + NaI	18	1.125
Au^+ doped KDP + NaI	17	1.0625

Optically transparent pure KDP and KDP crystals containing alkali halides with and without Au^+ ions were grown by slow evaporation technique. Elemental analysis confirms that the impurities have entered into the lattice of the KDP crystals. KDP crystals containing alkali halides have shown appreciable increase in SHG efficiency compared to pure KDP crystals. Enhanced efficiency confirms the better alignment of additives and dopants in the crystal matrix.

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